CLOSING REPORT

KKP129866 Frontiers project: Quantum-coherent materials

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Total NKFIH budget: 296,594,000 HUF

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Abstract: The research focuses on point defects in solids that may act as single photon source or perhaps quantum bits. The host crystals and point defects acting as single photon sources or quantum bits together are called quantum coherent materials. These can be employed in the development of quantum communication application and nanoscale sensors. The main goal of the project is to find prospective quantum coherent materials by means of systematic ab initio calculations that may open new doors to science and development of quantum information.

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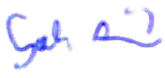

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Introduction

In this project, Ádám Gali's research group developed, implemented and employed *ab initio* methods to study the magneto-optical properties of point defects in solids that may act as quantum bits (see [Nature Reviews Materials 6, 906 (2021)] and references therein). Ádám Gali's team also applied group theory and numerical methods to analyze the electron-nuclear quantum systems and the spin-spin interactions that play a key role in the qubit's properties. The number of experimentally observed defect qubits was very limited around the start of the project [Applied Physics Reviews 7, 031308 (2020)]. The main goals of the project were to seek novel defect qubit candidates for a given quantum technology application and to optimize the quantum optics protocols of the known ones.

The investigations were often carried out in cooperation with world-leader experimental partners, where Prof. David D. Awschalom (U. Chicago), Prof. Mikhail D. Lukin (Harvard University), Prof. Nathalie P. de Leon (Princeton University), Prof. Shimon Kolkowitz (UC Berkeley), Dr. Jin-Shi Xu (U. Science and Technology of China), Prof. Jörg Wrachtrup (U. Stuttgart), Prof. Fedor Jelezko (U. Ulm), Prof. Milos Nesladek (U. Hasselt & IMEC), Dr. Michael Trupke (U. Wien), Prof. Vincent Jacques (U. Montpellier), Prof. Jean-François Roch (U. Paris-Saclay) and Dr. Thierry Debuisschert (Thales) partners are highlighted. During the project period, Ádám Gali was a PI in 7 European consortial projects (4 QuantERA, one H2020 and one HE projects).

Methods

We applied *ab initio* methods to calculate the electronic structure of color centers which is based on supercell plane wave density functional theory methods. The color centers were modeled by 512-atom or larger supercell. The spin-orbit and electron-phonon, and spin-spin interactions play a crucial role in the fine electronic structure of the defect qubits. In quantum sensor applications, the surface states can also play an important role that were studied with supercell slab models. A detailed review about these methods was written by Ádám Gali [Nanophotonics 8, 1907 (2019)]. In the calculation of the excited states, Ádám Gali's theory research group employed methods that were went beyond density functional theory, e.g. density matrix renormalization group in cooperation with Prof. Örs Legeza at the institute as summarized by Ádám Gali [Nanophotonics 12, 359 (2023)].

The calculations were carried out on massively parallelized computer clusters with the use of both local facilities, national (KIFÜ) and international supercomputer centers. The major part of investments (of about 48M HUF + VAT) was used in the first period of the project for increasing the computational capacity at the institute which was necessary to efficiently carry out the developments and implementation of novel methods.

Results

Ádám Gali's group has been achieved numerous results that can be found in the scientific publications. In the next sections, we highlight the most important results below. We note that some of the results not only recognized by the Editors of the journals as featured articles but the PI was asked to give interviews

in the media about the relevance of the results (e.g., https://www.klubradio.hu/archivum/otos-para-kovacs-imrevel-2022-majus-30-hetfo-1005-25369).

Optimization of quantum optics protocols for known defect qubits

Significant results have been achieved for the exemplary nitrogen-vacancy (NV) center in diamond. Understanding the ionization processes led to an increase in the optically detected magnetic resonance (ODMR) contrast recorded at room temperature which can be used to improve the sensitivity in quantum sensor applications. At cryogenic temperatures, a theory is developed for the nuclear spin flips upon illumination which can be used to employ a novel quantum optics protocol for increasing the sensitivity. A theory is developed and implemented at *ab initio* level to calculate the spin-phonon interaction and the respective longitudinal spin-relaxation rate as a function of temperature which could explain the double exponential feature of this function.

The coupling parameters of divacancy qubits in silicon carbide (SiC) were determined by *ab initio* calculations. Furthermore, divacancy-related qubits within cubic inclusions were proposed as the origin of the so-called PL5/6 centers in 4H-SiC that have as high room temperature ODMR contrast as the nitrogen-vacancy center in diamond. For the silicon-vacancy qubits in SiC, the nature of the optical excited state was found by our theory and *ab initio* calculations. The results suggested that the so-called V2 center in 4H-SiC is favored over V1-center (different forms of silicon-vacancy in 4H-SiC) for quantum communication applications.

Proposal for novel defect qubits

A strong electron-phonon coupling was found for the neutral excited state of group-IV-vacancy centers in diamond that was described by product Jahn-Teller interaction and parametrized by *ab initio* calculations. The role of bound exciton states for the neutral silicon-vacancy center in diamond was discovered in the ODMR observation for this qubit. The negatively charged nickel-related centers were identified and suggested as qubits for quantum communication.

Combined theoretical and experimental efforts identified the telecom emitters in 4H SiC which are two distinct configurations of the vanadium defect. The electronic structure and optical properties were analyzed by combination of group theory and first principles calculations.

Furthermore, we could identify potential qubits in silicon which is a very promising platform for quantum communication applications. The so-called G-center and W-center were identified and the so-called C-center was characterized in details emitting in the telecommunication L-band. For the G-center, a microscopic mechanism was identified for their creation which involves a substitutional carbon defect and a migrating interstitial carbon defect in silicon.

Low dimensional structures were also studied by Ádám Gali's group. The electrical and optical properties of boron phosphide nanoparticles were studied by time dependent density functional theory calculations. The quantum confinement regime was identified and its effect to the photocatalytic activity.

Highly correlated orbitals coupled with phonons in two-dimension were identified for paramagnetic and optically active boron vacancy in hexagonal boron nitride (hBN) by first principles methods which are responsible for recently observed optically detected magnetic resonance signal. *Ab initio* analysis of the

correlated electronic structure of this center by density matrix renormalization group and Kohn-Sham density functional theory methods were reported. By establishing the nature of the bright and dark states as well as the position of the energy levels, a complete description of the magneto-optical properties and corresponding radiative and non-radiative routes was provided which are responsible for the optical spin polarization and spin dependent luminescence of the defect. These findings pave the way toward advancing the identification and characterization of room temperature quantum bits in two-dimensional solids. Later, we simulated the complex interplay between the electron spin and nuclear spins for this defect that revealed the complex feature in the observed spin dynamics of the electron spin of boron-vacancy in hBN.

Ádám Gali's group studied the effect of strain on the physical properties of the nitrogen antisite-vacancy pair in hBN, a color center that may be employed as a quantum bit in a two-dimensional material. With group theory and ab-initio analysis it was shown that strong electron-phonon coupling plays a key role in the optical activation of this color center. A giant shift on the zero-phonon-line (ZPL) emission of the nitrogen antisite-vacancy pair defect was found upon applying strain that is typical of hBN samples. These results provide a plausible explanation for the experimental observation of quantum emitters with similar optical properties but widely scattered ZPL wavelengths and the experimentally observed dependence of the ZPL on the strain. Many quantum emitters have been measured close or near the grain boundaries of hBN where various Stone-Wales defects appear. It was shown by means of first principles density functional theory calculations that the pentagon-heptagon Stone-Wales defect is an ultraviolet emitter and its optical properties closely follow the characteristics of a 4.08-eV quantum emitter, often observed in polycrystalline hexagonal boron nitride. It was also shown that the square-octagon Stone-Wales line defects are optically active in the ultraviolet region with varying gaps depending on their density in hBN. Our results may introduce a paradigm shift in the identification of fluorescent centers in this material.

Furthermore, single spin ODMR centers were identified as special carbon defects in hBN which has been a breakthrough and published in a *Nature Materials* paper together with Jörg Wrachtrup group where the detailed theory results were also reported in a *Physical Review B* paper. The origin of the single photon emitters in the visible region was associated with oxygen-vacancy defects whereas the origin of the ultraviolet single photon emitters was associated with special carbon dimer and larger carbon clusters by *ab initio* calculations.

In addition, a potential qubit in WS_2 was proposed as the carbon substituting sulfur in the neutral charge state which should exhibit a near infrared emission in the telecom wavelength.

Outlook

Despite on the pandemic crisis in the year of 2020, significant results could be achieved. The results were published in prestigious journals, e.g. Nature Materials, Nature Reviews Materials, Nature Communications, Nature Partner Journal Computational Materials, Nature Partner Journal Quantum Information, Science Advances, National Science Review, Physical Review Letters, Nano Letters, ACS Nano and other prestigious journals (total impact factor of ~675 with 75 papers). A few papers were selected by Editors and they put those into spotlight. The published papers already received ~1100 independent citations according to MTMT (Hungarian scientific database on 2022-12-31) and attracted great attention in the community of quantum technology researchers. The results were also presented in contributed and

invited talks at workshops and conferences. It is noted that because of the pandemic crisis the conference invitations were naturally ceased in 2020. The invited talks are listed below:

- *Basic properties and quantum control of diamond qubits*, Quantum Innovation 2023, Tokyo (Japan), November 15-17, 2023

- *Theory of quantum bits in diamond*, "Quantum-coherent structures: Quantum computing" Workshop, University of Leipzig, Germany, September 28-29, 2023

- *Theoretical magneto-optical spectroscopy of solid state defect quantum bits*, Eddleman Quantum Institute (EQI) Workshop, Chateau de Balleroy, France, September 7-10, 2023

- *Quantum sensors from nitrogen-vacancy center in diamond: theory*, RAISIN 2023, Guildford (UK), September 6-8, 2023

- *Theoretical magneto-optical spectroscopy of solid state defect quantum bits*, Theory of Condensed Matter Seminar at Cambridge University, Cambridge (UK), June 8, 2023

- *Defect spins and qubits in hBN from first principles theory guiding experiments*, BNW2023 - Workshop on Boron Nitride, Montpellier (France), May 29 - June 2, 2023

- *Control of near-surface nitrogen-vacancy quantum sensor in diamond*, Trends in Quantum Matter, Stavropoulos Center for Quantum Matter at University of Notre Dame, Notre Dame (USA), May 18-19, 2023

- *Defect spins and qubits in hexagonal boron nitride from first principles theory guiding experiments*, Defects in Two-dimensional Materials - 750. WE-Heraeus-Seminar, Bad Honnef (Germany), May 8-12, 2023

- *Ab initio theory of colour centres in diamond*, Theo Murphy Conference - Diamond for Quantum Applications, October 10-11, 2022, Whittlebury Park (UK)

- *Rejuvenation of the magneto-optical spectroscopy of silicon defects: promising single photon sources and qubits*, European Materials Research Society Fall Meeting 2022, 19-22 September, 2022, Warsaw (Poland)

- *Theoretical magneto-optical spectroscopy for solid state defect qubits*, 50th International School and Conference on the Physics of Semiconductors, "Jaszowiec 2022", Szczyrk (Poland), 4-10 June, 2022

- *Near-infrared wavelength quantum emitters*, Materials Research Society Fall Meeting 2021, Boston (USA), November 30 - December 3, 2021

- *Inversion symmetric color centers in diamond for quantum technology*, Materials Research Society Fall Meeting 2021, Boston (USA), November 30 - December 3, 2021

- *Ab initio theory of NV center and other centers*, Tutorial lecture, ASTERIQS School on Solid-State Spins, Cargèse, Corsica (France), November 2, 2021

- *Solid state defect qubits - toward full ab initio description*, Tutorial lecture, 31st International Conference on Defects in Semiconductors, Oslo (Norway), July 26, 2021

- *The theory of solid state defect qubits*, Computational Atomic-scale Materials Design (CAMD) webinar, Technical University of Denmark (DTU) Physics, March 8, 2021

- *First principles calculation of highly anisotropic g-tensor of Kramers doublet transition metals in hexagonal SiC*, International Conference on Silicon Carbide and Related Materials 2019, Kyoto (Japan), September 28 - October 4, 2019

- *Novel color centers in diamond for communication and sensing*, European Materials Research Society Fall Meeting 2019, Warsaw (Poland), September 16-19, 2019

- *Theory of silicon-vacancy and related colour centres in diamond*, 30th International Conference on Diamond and Carbon Materials, Sevilla (Spain), September 10-14, 2019

- *The progress in ab initio description of solid state defect qubits*, 30th International Conference on Defects in Semiconductors, Seattle (United States), July 21-26, 2019

- *Ab initio study of defect qubits for hyperpolarization and quantum sensing*, Gordon Research Conference on Quantum Sensing, Hong Kong (China), June 2-6, 2019

- *Ab initio study of defect qubits for hyperpolarization, quantum sensing and communication*, QDiamond Workshop, Tel Aviv (Israel), May 13-16, 2019

- *Theory of electrical readout of deep defect qubits in solids*, APS March Meeting, Boston (USA), March 4-8, 2019

Young researchers were trained and worked in the project. András Csóré, Gyula Károlyházy and Péter Udvarhelyi started to work as a PhD candidate on the project, and they could successfully defend their PhD thesis during the project period. Viktor Ivády worked as an experienced postdoc researcher in the beginning of the project and he then got a Lendület award in 2022 from the Hungarian Academy of Sciences and he has founded his own research group at Eötvös Loránd University. András Csóré, Dávid Beke, and Gergő Thiering were decorated by scientific awards, e.g. Junior Príma and Róbert Bárány awards.

As a consequence of the results achieved by Ádám Gali's group, new joint projects have been started with forming European research consortium in the field of quantum technology.